

Particle displacements in the elastic deformation of amorphous materials: local fluctuations vs. non-affine field

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Abstract. - We study the local disorder in the deformation of amorphous materials by decomposing the particle displacements into a continuous, inhomogeneous field and the corresponding fluctuations. We compare these fields to the commonly used non-affine displacements, in an elastically deformed 2D Lennard-Jones glass. Unlike the non-affine field, the fluctuations are very localized, and exhibit a much smaller (and system size independent) correlation length, on the order of a particle diameter, supporting the applicability of the notion of local “defects” to such materials. We propose a scalar “noise” field to characterize the fluctuations, as an additional field for extended continuum models, e.g., to describe the localized irreversible events observed during plastic deformation.

Introduction. – The nature of fluctuations in glasses and other amorphous materials out of equilibrium is of much current interest. While elasticity and plasticity are often employed for describing both crystalline and amorphous materials, their microscopic basis is well-established only in crystalline (or polycrystalline) materials, and relies on the periodicity of the microscopic structure (possibly with localized defects) [1, 2]. As in crystal plasticity, localized rearrangements appear to play an important role in the plastic deformation of amorphous materials [3–5], but the lack of underlying order renders the identifications of localized “defects” difficult. In crystals (with a simple unit cell) under homogeneous deformation, the particle displacements conform to the imposed (affine) strain, but in amorphous materials they do not [6]. The *non-affine* displacements (obtained by subtracting the expected homogeneous deformation) have recently been studied in experiments and simulations of different amorphous systems (e.g., glasses, colloids, granular materials and foams [7–14]). They are typically of the same order of magnitude as the relative affine displacements of neighbor-

ing particles, and therefore cannot be considered a small correction: ignoring them, or treating them as a perturbation, yields highly inaccurate estimates for macroscopic material properties such as the elastic moduli [8, 15, 16]. Considering the non-affine displacements as a fluctuation, or “noise” [8, 10], poses difficulties since they exhibit long range correlations [8, 10–13, 17] which would render the contribution of such “noise” dominant at large scale.

In this Letter we show, using numerical simulations of a two dimensional (2D) Lennard-Jones glass subject to small elastic deformation, that the main features of the non-affine field can be captured by a continuous, inhomogeneous (subsystem scale) displacement field. However, the microscopic displacements exhibit significant fluctuations with respect to this field. A full characterization of the displacements therefore requires not only a distinction between an affine and non-affine contribution, but also between a continuous and a fluctuating part. We present the first study of the local fluctuations, and show that their properties are very different from those of the non-affine field: they are essentially uncorrelated and extremely localized; their distribution is qualitatively different. Furthermore, unlike the non-affine displacement, whose correlation depends on the system size (see [13] and below),

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the (very short) correlation of the fluctuations does not. We propose a new continuum scalar “noise” field based on the fluctuations. This field is formally analogous to the kinetic temperature in the kinetic theory of gases, which quantifies the local deviations from the coarse grained (hydrodynamic) velocity field. The “noise” field may be used for supplementing the macroscopic displacement field in extended continuum descriptions, e.g., to account for the part of the microscopic elastic energy not captured by the macroscopic field [18,19]. The localized nature of the fluctuations, and patterns observed in the “noise” field parallel to the principal shear directions, suggest a relation (confirmed by preliminary results [20]) between the fluctuations in the elastic response and irreversible localized rearrangements observed in plastic deformation [4,5].

Definitions. — Consider particles (labeled by Roman indices) whose center of mass positions are denoted $\{\mathbf{r}_i^0\}$ in a reference configuration and $\{\mathbf{r}_i\}$ in a deformed one. The displacement of particle i is $\mathbf{u}_i \equiv \mathbf{r}_i - \mathbf{r}_i^0$. In a simple periodic lattice (with one particle per unit cell), under a uniform strain ϵ , the deformation is *affine*, i.e., the relative displacement of two particles, $\mathbf{u}_{ij} = \mathbf{u}_i - \mathbf{u}_j$, is given by:

$$u_{ij\alpha} = \epsilon_{\alpha\beta} r_{ij\beta}^0, \quad (1)$$

where $\mathbf{r}_{ij}^0 = \mathbf{r}_i^0 - \mathbf{r}_j^0$ and Greek indices denote Cartesian coordinates (the Einstein summation convention is used). This relation between the particle displacements and the macroscopic deformation forms the microscopic basis of crystal elasticity [1]. It has long been appreciated [6] that in disordered systems, the displacements are not affine even under uniform applied strain, since an affine deformation would result in a configuration in which force balance is violated. The non-affine particle displacements,

$$\delta u_{i\alpha} \equiv u_{i\alpha} - \epsilon_{\alpha\beta} r_{i\beta}^0, \quad (2)$$

typically exhibit long range correlations [8,10,11,13,17], which depend on the system size (as observed in [13] and discussed further below): the correlation of their scalar product crosses zero at a distance of about 0.3 times the system width. This can be explained by treating the material as inhomogeneously elastic, which predicts a power law decay of the correlations (logarithmic in 2D) [12,13]. Therefore, rather than treating the non-affine displacements as discrete, particle scale fluctuations, their main features may be captured by an inhomogeneous, subsystem scale continuum description.

In order to define microscopic displacement fluctuations, one needs to subtract a local, continuous displacement field from the individual particle displacements. Rather than performing an arbitrary smoothing or interpolation of the particle displacements, we employ a definition proposed in [18], based on a systematic spatial coarse graining (CG) procedure. This procedure uses the standard definition of the microscopic densities of mass, momentum and energy as sums over Dirac delta functions centered at the particles’ centers of mass. The corresponding

CG densities are obtained by a convolution with a spatial CG function $\phi(\mathbf{R})$, a normalized non-negative function with a single maximum at $\mathbf{R} = 0$ and a characteristic width w , the CG scale (e.g., a Gaussian: in 2D, $\phi(\mathbf{R}) = \frac{1}{\pi w^2} e^{-(|\mathbf{R}|/w)^2}$). In particular, the CG mass density is $\rho(\mathbf{r}, t) \equiv \sum_i m_i \phi[\mathbf{r} - \mathbf{r}_i(t)]$, and the CG momentum density is $\mathbf{p}(\mathbf{r}, t) \equiv \sum_i m_i \mathbf{v}_i(t) \phi[\mathbf{r} - \mathbf{r}_i(t)]$, where $\{m_i\}, \{\mathbf{v}_i(t)\}$ are the particle masses and velocities at time t , respectively. The definition of the CG displacement follows its standard definition in continuum mechanics, i.e., the Lagrangian time integral over the velocity of a *material particle*. The CG velocity is given by:

$$\mathbf{v}(\mathbf{r}, t) \equiv \frac{\mathbf{p}(\mathbf{r}, t)}{\rho(\mathbf{r}, t)} = \frac{\sum_i m_i \mathbf{v}_i(t) \phi[\mathbf{r} - \mathbf{r}_i(t)]}{\sum_j m_j \phi[\mathbf{r} - \mathbf{r}_j(t)]}. \quad (3)$$

Therefore, the CG displacement field is given by [18,21]:

$$\begin{aligned} \mathbf{U}(\mathbf{R}, t) &\equiv \int_0^t \mathbf{v}[\mathbf{r}(\mathbf{R}, t'), t'] dt' \\ &= \int_0^t \frac{\sum_i m_i \mathbf{v}_i(t') \phi[\mathbf{r}(\mathbf{R}, t') - \mathbf{r}_i(t')]}{\sum_j m_j \phi[\mathbf{r}(\mathbf{R}, t') - \mathbf{r}_j(t')]} dt' \\ &= \frac{\sum_i m_i \mathbf{u}_i(t) \phi[\mathbf{R} - \mathbf{r}_i^0]}{\sum_j m_j \phi[\mathbf{R} - \mathbf{r}_j^0]} + \mathcal{O}(\epsilon^2), \end{aligned} \quad (4)$$

where \mathbf{R} denotes the Lagrangian coordinate of a material particle whose (Eulerian) coordinate at time t is \mathbf{r} , $\mathbf{u}_i(t)$ is the total displacement of particle i , and ϵ is a measure of the local strain; the last line is obtained using integration by parts [18]. The particle masses in Eq. (4) ensure consistency with a dynamical (time dependent) description [18]; for the quasi-static deformation considered here, t may be interpreted as a parameter characterizing the overall deformation. For small strain, our focus in this Letter, one can use the approximate expression on the last line of Eq. (4), which is trajectory independent, i.e., it involves only the total particle displacements. With a smooth $\phi(\mathbf{R})$, Eq. (4) defines a smooth displacement field which can be used to calculate local, scale dependent strain measures based on its gradients. Unlike strain definitions based on a local fit to an affine deformation (e.g., [3,16]), those are, by construction, fully consistent with continuum mechanics, and may be extended to large deformations. The CG displacement is defined only in terms of the particle displacements, and does not rely on any assumptions regarding material homogeneity or constitutive description, as does the decomposition into an affine and non-affine part. It can therefore also be used for describing inelastic deformation (e.g., plastic flow or fracture). Based on Eq. (4), we define the *displacement fluctuations* as $\mathbf{u}'_i(\mathbf{r}) \equiv \mathbf{u}_i - \mathbf{U}(\mathbf{r})$. For comparison with the non-affine field, it is useful to consider the fluctuations at the particle positions, $\mathbf{u}'_i(\mathbf{r}_i^0)$, denoted below as \mathbf{u}'_i . The CG displacement field, $\mathbf{U}(\mathbf{r})$, and hence the fluctuations \mathbf{u}'_i , are in general *scale dependent*. A possible definition of a corresponding CG scalar “noise” field is $\eta(\mathbf{r}) \equiv \sum_i m_i |\mathbf{u}'_i(\mathbf{r})|^2 \phi[\mathbf{r} - \mathbf{r}_i(t)]$. This expression is formally similar to the kinetic temperature in

the kinetic theory of gases [19] with the velocity fluctuations replaced by the displacements fluctuations (except for a density factor; the density is quite homogeneous in the systems considered here).

Results. — We apply the above definitions to 2D amorphous solids [8, 10] prepared by quenching a fluid of polydisperse particles (with diameters $\{\sigma_i\}$ uniformly distributed in the range $0.8\sigma - 1.2\sigma$) of equal (unit) mass, interacting via a Lennard-Jones potential, $V_{ij}(r) = 4\epsilon [(\sigma_{ij}/r)^{12} - (\sigma_{ij}/r)^6]$, where $\sigma_{ij} = (\sigma_i + \sigma_j)/2$. The mean particle diameter (σ) defines the unit of length. We use periodic boundary conditions in both directions. We apply three different uniform deformation modes: uniaxial stretching parallel to the x or y axis, and a simple shear parallel to the x axis. For a uniaxial deformation, the length of the simulation cell is multiplied by $1 + \epsilon$, while for simple shear, we use Lees-Edwards boundary conditions with shear strain $\gamma = \epsilon/2$. We impose the affine displacement [Eq. (1)] on each particle, and subsequently relax the system to the nearest energy minimum [11]. We use $\epsilon = 10^{-6}$, which ensures a linear response [5]. Unless noted otherwise, the data presented were obtained using systems with $N = 10000$ particles of size 104×104 (i.e., the mean density is $\rho = 0.925$).

Figs. 1a,b show the non-affine particle displacements, $\delta\mathbf{u}_i$, in one configuration under simple shear. The vortex-like structures are qualitatively similar to those observed in simulations and experiments on amorphous materials (e.g., [7, 9–11, 17, 22]). The corresponding displacement fluctuations, \mathbf{u}'_i , calculated using a Gaussian CG function with $w = 1$, are shown in Figs. 1c,d (the choice of w is discussed below). The fluctuations exhibit quite a different behavior from the non-affine field: they are more localized and considerably less correlated (as discussed below). The degree of localization can be quantified by the participation ratio, defined as $p \equiv (\sum_i |\mathbf{u}'_i|^2)^2 / [N \sum_j (|\mathbf{u}'_j|^2)^2]$. Averaging over an ensemble of 3 independent deformation modes for each of 9 different realizations, we obtain (with $w = 1$) $p_w = 0.08 \pm 0.02$, while for the non-affine field $p_{\delta u} = 0.28 \pm 0.06$ (the error is the standard deviation in the ensemble); the same ensemble is used for Figs. 3, 5 and 6 below. The mean magnitude of the fluctuations is also smaller, by a factor of about 4 ($4.4 \cdot 10^{-7}$ vs. $1.7 \cdot 10^{-6}$).

The “noise” field, $\eta(\mathbf{r})$, presented in Fig. 2, shows very clearly the localized nature of the fluctuations. It exhibits rather sharp peaks, whose positions depend both on the configuration and on the applied deformation (e.g., shear or stretching). Preliminary results [20] indicate that these peaks are correlated with the positions of localized plastic rearrangements observed in plastic deformation [5]. Plotting the logarithm of the noise (Figs. 2b,d) reduces the contrast and reveals anisotropic patterns oriented predominantly near the principal shear directions (parallel to the axes for simple shear, Fig. 2b, and at 45° to the axes for uniaxial stretching, Fig. 2d). These correspond to the typ-

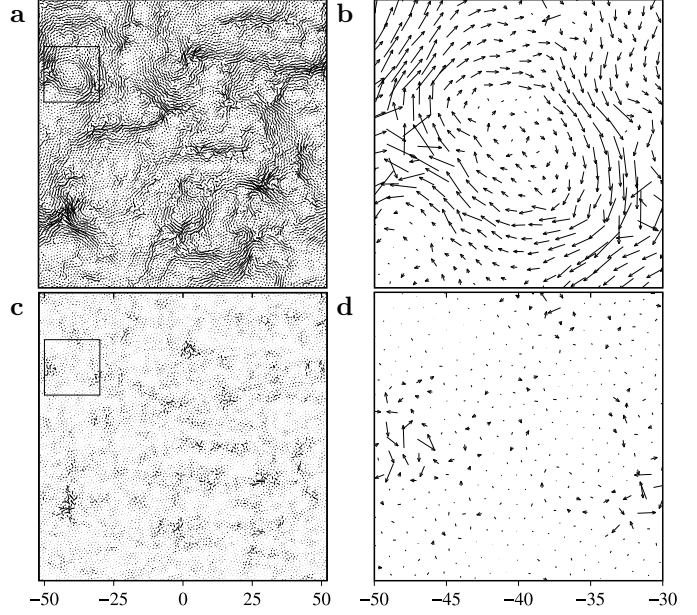


Fig. 1: a. the non-affine displacements, $\delta\mathbf{u}_i$; c. the displacement fluctuations, \mathbf{u}'_i (with CG width $w = 1$) in a 10000 particles 2D polydisperse Lennard-Jones glass under simple shear $\gamma = 5 \cdot 10^{-7}$ (all vectors are magnified by $5 \cdot 10^5$); b,d. a corresponding zoom on a region of size 20×20 particles.

ical directions of the localized shear bands observed in the plastic regime [5], which provides further support to the notion that fluctuations and inhomogeneities in the elastic regime are related to subsequent plastic failure and flow.

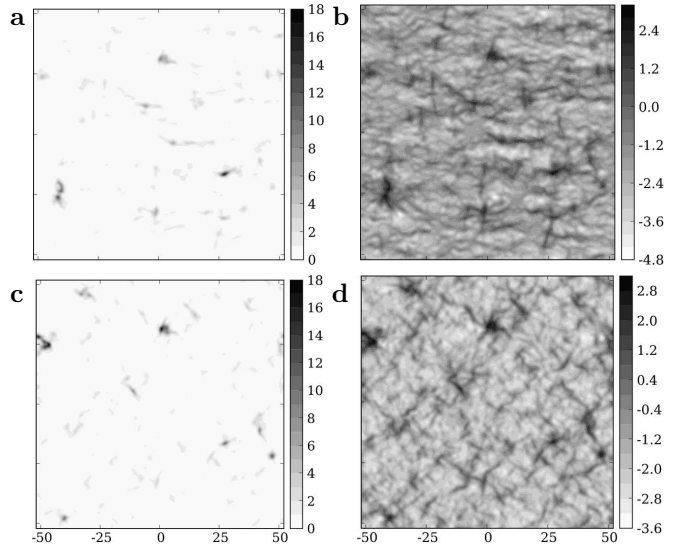


Fig. 2: Contour plots of (a,c) the noise field, $\eta(\mathbf{r})$ (with $w = 1$), in the system shown in Fig. 1, scaled by $2.5 \cdot 10^{11}$ and (b,d) its logarithm, for two different applied deformations: simple shear (a,b) and uniaxial stretching parallel to the x -axis (c,d). The noise field is calculated on a grid whose spacing is $w/4 = 1/4$.

Another striking difference between the displacement fluctuations and the non-affine

field is observed in their correlation. Fig. 3 shows the scalar product correlation function, $C(R) = \sum_{ij} \mathbf{u}_i \cdot \mathbf{u}_j \delta(R - |\mathbf{r}_i - \mathbf{r}_j|) / \sum_{ij} \delta(R - |\mathbf{r}_i - \mathbf{r}_j|)$, normalized by its value at $R = 0$ (i.e., the mean squared vector magnitude) for the three fields. The correlation of the non-affine field appears to be logarithmic on intermediate scales. This behavior is captured quite well by the CG displacement with $w = 1$. The characteristic scale for the correlation of the non-affine field (for this system size, 104×104) is about 30, similar to the diameter of the vortices (Fig. 1), while for the displacement fluctuations it is on the order of one diameter. This justifies the assumption, made implicitly in [18], of the absence of long range correlations in the fluctuations, required for demonstrating that *local* linear elasticity applies on scales sufficiently large compared to their correlation length.

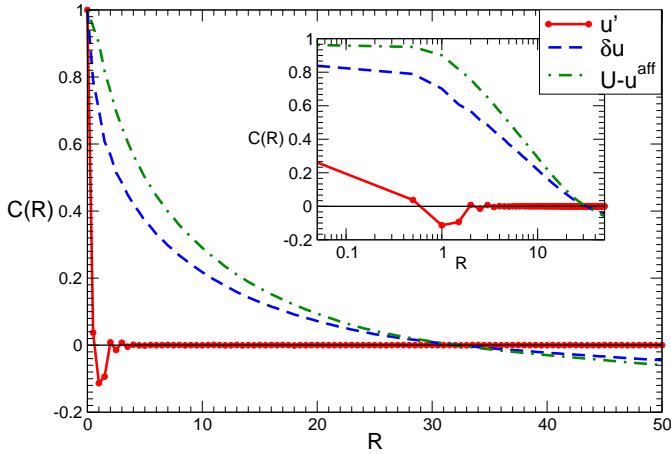


Fig. 3: Main: ensemble averaged correlation of the scalar product of the non-affine displacements, $\delta \mathbf{u}_i$, the CG displacements at the particle positions, $\mathbf{U}(\mathbf{r}_i^0)$ (with $w = 1$, the affine part subtracted) and the displacement fluctuations, \mathbf{u}'_i ; inset: same graph with logarithmic horizontal scale.

As mentioned, the correlation of the non-affine field has been observed to depend on the system size, in a somewhat different system of bidisperse disks with harmonic repulsion [13]. We verified that this is indeed the case in the systems we consider here: Fig. 4 presents the scalar product correlation function of the non-affine field and of the displacement fluctuations for four system sizes: 46.5×46.5 ($N = 2000$), 104×104 ($N = 10000$), 208×208 ($N = 40000$) and 483.5×483.5 ($N = 216225$), a considerably larger range of sizes than considered in [13]. The results were obtained by averaging over 20, 8 and 8 different realizations respectively for the first three sizes (only one configuration with $N = 216225$ was used), for one deformation mode: uniaxial stretching parallel to the x axis; representative error bars indicate the standard deviation in the ensemble. The correlation of the non-affine field exhibits a rather good collapse (at least for the larger sys-

tems considered) when the distance is scaled by the length of the system, L (the deviation observed for $N = 216225$ may be due to reduced statistics, or to two weak “plastic events” which occurred in the system, indicating a possible deviation from a linear response). On the other hand, as shown in the inset of Fig. 4, the correlation of the displacement fluctuations does not depend on the system size, i.e., unlike the non-affine field, it provides a *local* characterization of the disorder in the particle displacements.

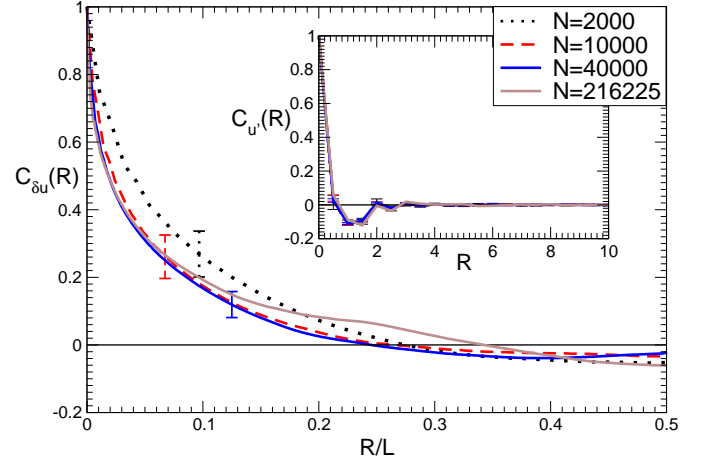


Fig. 4: Main: ensemble averaged correlation of the scalar product of the non-affine displacements, $\delta \mathbf{u}_i$, for systems of different sizes, subject to uniaxial stretching parallel to the x axis, with the distance R scaled by the length of the system, L ; inset: the correlation of the displacement fluctuations, \mathbf{u}'_i (with $w = 1$), in the same systems, distance unscaled.

The CG displacement and its fluctuations obviously depend on the chosen CG scale (resolution), w . For $w \rightarrow 0$, $\mathbf{U}(\mathbf{r}_i^0) = \mathbf{u}_i$ so that $\mathbf{u}'_i = 0$. For $w \rightarrow \infty$, $\mathbf{U}(\mathbf{r}_i^0) = 0$, since the center of mass of the system is fixed, hence $\mathbf{u}'_i = \mathbf{u}_i$. In order to obtain the optimal continuum description we would like to choose w to be as small as possible. In lattices with a complex unit cell, the deformation can only be expected to be locally homogeneous, or affine, on scales larger than the size of the unit cell. In our case there is no periodicity in the structure; hence, by analogy, the minimum scale on which we can expect to define a meaningful CG displacement is on the order of the interparticle separation, $w \simeq \langle \sigma \rangle = 1$. The oscillatory nature of the correlation of the scalar product of \mathbf{u}'_i (Figs. 3,4) makes it difficult to define a correlation length; we therefore use the correlation of the *magnitudes*, $|\mathbf{u}'_i|$, which is fit rather well by $C_{|\mathbf{u}'|}(R) = \exp(-R/\xi_{|\mathbf{u}'|})$, at least for sufficiently small w ; see the inset of Fig. 5. In Fig 5, we present the dependence on w of the participation ratio, $p_{u'}$, and of the correlation length $\xi_{|\mathbf{u}'|}$. As expected, the results are scale dependent: the goal is to characterize the *inhomogeneity* of the displacement field in particular configurations; scale independence is expected only for locally homo-

neous fields, or for ensemble averages when such averages are homogeneous [23]. The participation ratio increases sharply with w up to $w \simeq 1$, above which it increases more slowly, eventually reaching (for large w ; not shown) values similar to those of the non-affine field, as expected. The correlation length, $\xi_{|u'|}$, shows a small “plateau” around $w \simeq 1$, at a value of about 1.5 mean diameters. This supports our choice of $w = 1$ for separating the smooth, continuous part of the displacement from the “noise”. As expected, $\xi_{|u'|}$ increases with w (due to the long range correlation of the CG displacement, hence the reduced quality of the exponential fit), but remains much smaller than the typical length observed for the non-affine field (Figs. 3,4).

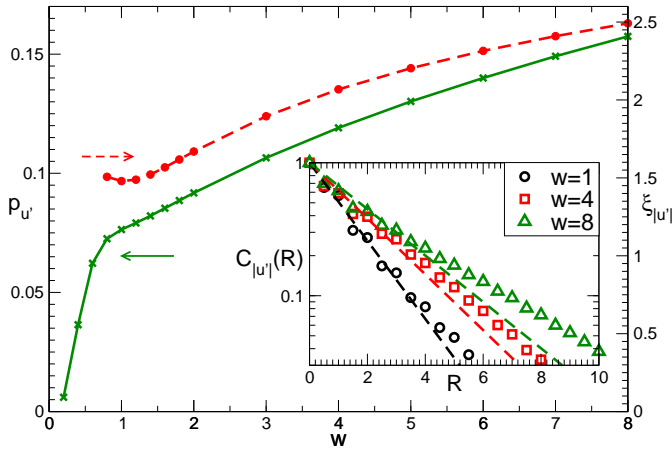


Fig. 5: Main: correlation length, $\xi_{|u'|}$, and participation ratio, $p_{u'}$, of the displacement fluctuations, \mathbf{u}'_i , vs. the CG width, w ; inset: ensemble averaged correlation of the magnitude of the displacement fluctuations, $|\mathbf{u}'_i|$, for three values of w (symbols); dashed lines indicate the exponential fits used to calculate $\xi_{|u'|}$.

It is interesting to compare the ensemble averaged distributions of the components of the displacement fluctuations (with $w = 1$) and of the non-affine field, shown in Fig. 6 (averaged over the x and y components, whose distributions are the same within statistical error, i.e., they are isotropic). The two distributions are qualitatively different. In particular, the center of the distribution is exponential for the fluctuations (the fit shown in Fig. 6 is for $\tilde{u}'_{i\alpha} < 3.5$), but Gaussian (the fit shown is for $\delta\tilde{u}_{i\alpha} < 1.6$) for the non-affine field (see also [9]). Both distributions exhibit power law tails, with different exponents. For $w \gtrsim 1$, we find (not shown) that the distribution of the fluctuations crosses over to a Gaussian at the center, presumably due to the contribution of the continuous part of the displacement (which is dominant in the distribution of the non-affine field). This provides further support for the choice of $w = 1$. Note that the crossover to a Gaussian distribution is not due to the Gaussian CG function used: a Heaviside CG function yields similar results.

In [8] (Fig. 10), a length $\xi \simeq 30 \langle \sigma \rangle$, independent of

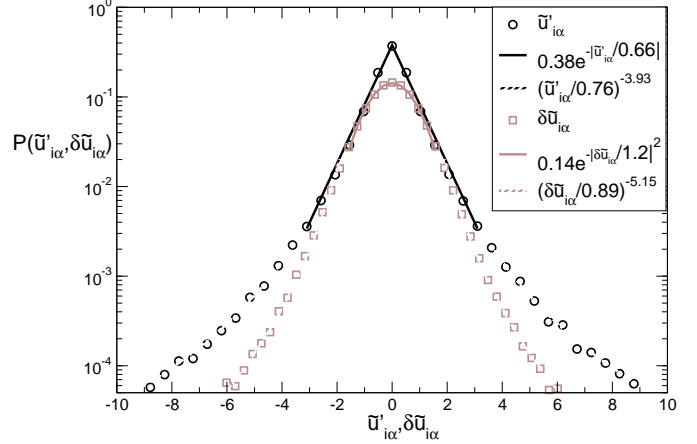


Fig. 6: Ensemble averaged distributions of both components of the non-affine displacement, $\delta\tilde{u}_{i\alpha} \equiv \delta\mathbf{u}_{i\alpha}/\sigma_{\delta u}$, and of the displacement fluctuations, $\tilde{u}'_{i\alpha} \equiv \mathbf{u}'_{i\alpha}/\sigma_{u'}$ (with $w = 1$), where $\sigma_{\delta u} = 1.31 \cdot 10^{-6}$ and $\sigma_{u'} = 4.05 \cdot 10^{-7}$ are the corresponding standard deviations.

the system size, was found to characterize the crossover of the vibrational modes to their continuum limit, in systems similar to the ones studied here. A similar crossover length seems to apply for a uniform applied strain [8,18]. It is not clear how, and whether, this length is related to the disorder in the displacement field. The correlation lengths of the fluctuations (Fig. 4), as shown above, or of the noise field (about 2.1 with $w = 1$), are much smaller than ξ , and are system size independent; however, the correlation of these fields does not clearly reveal an *additional* length. The *distribution* of the fluctuations (or noise) is also essentially size independent: Fig. 7 presents the distribution of the noise field for systems of different sizes (averaged over an ensemble of configurations, except for the larger system; the deviations observed for the latter are probably due to the “plastic events” mentioned above, around which the noise is larger). We verified the size independence of the distributions of the fluctuations and of the non-affine field. The enhancement of small noise for the sheared systems is also observed in the components of \mathbf{u}'_i , for which the probability is larger by about 10% for $\tilde{u}'_{i\alpha} \lesssim 0.2$; this effect is too small to be seen in Fig. 6, which justifies the averaging over deformation modes used in that figure. The patterns observed in Figs. 2b,d probably reflect a more significant difference in the *correlation* for different deformation modes rather than this small difference in the distribution. Defining a density of “defects”, i.e., regions of large noise (the peaks visible in Figs. 2a,c) would enable the definition of a corresponding length. However, the objective identification of the defects is difficult, since the power law decay of the noise distribution precludes the definition of a natural cutoff for the noise. We have therefore not been able to clearly identify the mesoscopic length scale ξ in the fluctuations (or the non-affine field).

This remains an important issue for further study.

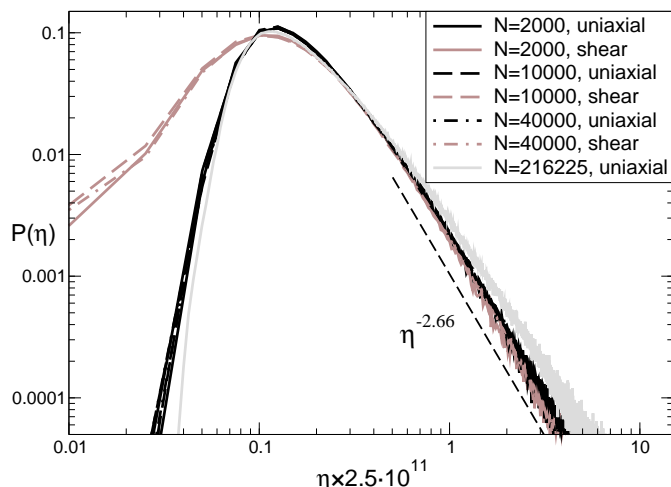


Fig. 7: Ensemble averaged distributions of the noise field, $\eta(\mathbf{r})$ (with $w = 1$), scaled by $2.5 \cdot 10^{11}$, in systems of different size, subject to either uniaxial stretching or simple shear.

Conclusion. — Our results suggest that the main features of the non-affine field may be described by an inhomogeneous continuum model, e.g., local linear elasticity (as in [12]) which should apply on sufficiently large scales, on the order of ten diameters [8, 10, 11, 18]; however, corrections to local elasticity are expected to be required on smaller scales. Both the non-affine field and the “intrinsic” microscopic fluctuations exhibit wide tailed distributions; however, the latter exhibit much shorter correlations. While the fluctuations cannot be described by classical continuum models, their short correlation should facilitate a statistical description, possibly in terms of local “defects”. We propose a related “noise” field which may be used for extending such models, e.g., to describe the part of the elastic energy which is not captured by the CG displacement [18, 19]. The localized nature of the “noise” field, as well as the patterns it exhibits, suggest a relation to the localized plastic rearrangements observed in plastic deformation [5]. This relation is confirmed by preliminary results [20], and is currently being studied in further detail. Such a relation may provide a (currently lacking) microscopic basis for phenomenological models for plastic flow involving localized rearrangements of zones which interact via the elastic deformation of the material, such as the Shear Transformation Zone (STZ) model [3] and similar models (e.g., [24, 25]). Both inhomogeneities (pertaining to the continuum description) and microscopic fluctuations may be important in this context. The approach suggested here should be useful both for analyzing simulations, as described in this Letter, and experimental data.

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